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Radionuclide Decay and In-growth Technical Basis Document

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Executive Summary

The purpose of this report is to assess the decay and in-growth of radionuclides from the radionuclide source term (RST) deposited by underground nuclear weapons tests conducted at the NTS from 1951 through 1992. A priority of the Underground Test Area (UGTA) project, administered by the Environmental Restoration Division of NNSA/NV, was to determine as accurately as possible a measure of the total radionuclide inventory for calculation of the RST deposited in the subsurface at the Nevada Test Site (NTS). The motivation for the development of a total radionuclide inventory is driven by a need to calculate the amount of radioactivity that will move away from the nuclear test cavities over time, referred to as the hydrologic source term (HST). The HST is a subset of the RST and must be calculated using knowledge of the geochemistry and hydrology of the subsurface environment. This will serve the regulatory process designed to protect human health from exposures to contaminated groundwater.

Following the detonation of an underground nuclear test, and depending on the presence of water at the location of the detonation, the residual radionuclides may be found in aqueous or gaseous states, precipitated or chemically sorbed states, or incorporated in melt glass produced by the nuclear test. The decay and in-growth of radionuclides may have geochemical implications for the migration of radionuclides away from underground nuclear test cavities. For example, in the case of a long-lived mobile parent decaying to a shorter-lived and less mobile daughter, the geochemical properties of the parent element may control the migration potential of the daughter nuclide. It becomes important to understand the evolution of the RST in terms of effects on the mobility, solubility, or abundance of radionuclides in the HST that are created by decay and in-growth processes.

The total radionuclide inventory and thus the RST changes with time due to radioactive decay. The abundance of a specific radionuclide at any given time is a function of the initial amount of radioactivity, the decay rate and in-growth from parent radionuclides. The in-growth of radioactivity is the additional amount of radioactivity for a given radionuclide that comes from the decay of the parent isotopes. In this report, decay and in-growth of radionuclides from the RST are evaluated over the 1000-year time frame in order to determine whether coupled in-growth and decay affect the relative abundance of any RST radionuclide. In addition, it is also necessary to identify whether any new derivative radionuclides not initially produced by the nuclear test but exist now as a result of in-growth from a parent radionuclide

One of the major goals of this report is to simplify the transport modeler's task by pointing out where in-growth is unimportant and where it needs to be considered. The specific goals of this document are to evaluate radionuclide decay chains and provide specific recommendations for incorporating radionuclide daughters of concern in the calculation of the radionuclide inventory. To this end, the following recommendations are given:

- All of the radionuclides in the radionuclide inventory with low to intermediate atomic numbers ($Z<80$) will decay directly into stable, nonradioactive

daughter products. They will not produce additional, derivative members of the radionuclide inventory.

- The short-lived radionuclides in decay chains with long-lived parents will exist in secular equilibrium with their parents and their abundances can be estimated from the abundance of their parent. Inclusion of the daughters as additional, derivative members of the radionuclide inventory is not necessary.
- There are daughters in the actinide chains, such as isotopes of radium and radon that are of concern in the environment, but the background from the natural uranium dwarfs the RST concentrations. Therefore, they do not need to be included as additional derivative members of the radionuclide inventory.
- There are eight decay chains that involve long decay and in-growth patterns that have been discussed in this report.
  - $^{93}Zr \rightarrow {}^{93m}Nb \rightarrow {}^{93}Nb$: The in-growth of ^{93m}Nb from ^{93}Zr over 1,000 years will not be significant. Over a 1000-year time frame all of the ^{93m}Nb will have decayed to stable ^{93}Nb, and therefore is not an additional, derivative radionuclide.
  - $^{150}Eu \rightarrow {}^{150}Gd \rightarrow {}^{146}Sm$: Over the 1000 year-time frame all of the ^{150}Eu will decay to ^{150}Gd. ^{150}Gd is an additional derivative radionuclide to the radionuclide inventory of Bowen et al. (2001). Gd-150 has such a long half-life, that over a 1,000-year analysis the daughter radionuclides can be effectively ignored in 1000-year decay and transport calculations.
  - $^{152}Eu \rightarrow {}^{152}Gd$: Over the 1000 year-time frame a fraction of the ^{152}Eu will decay to ^{152}Gd. ^{152}Gd is an additional derivative radionuclide to the radionuclide inventory of Bowen et al. (2001). Gd-152 has such a long half-life, that over a 1,000-year analysis the daughter radionuclides can be effectively ignored in 1000-year decay and transport calculations.
  - $^{238}Pu \rightarrow {}^{234}U$: The ^{238}Pu will gradually decay to ^{234}U in the 1000 years. This in-growth of ^{234}U will add to the ^{234}U from decay of ^{238}Pu and should be treated explicitly. This may be of consequence in tests with little ^{234}U. Further daughter products of ^{234}U should be insignificant over 1000 years and do not represent additional, derivative members of the radionuclide inventory.
  - $^{241}Pu \rightarrow {}^{241}Am \rightarrow {}^{237}Np$: This is probably the most important decay and in-growth chain in the RST. In-growth is important for calculating both ^{241}Am and ^{237}Np. The ^{241}Pu ($t_{1/2}$ = 14.4 y) will significantly increase the activity of the ^{241}Am and consequently the ^{237}Np on the 1000-year time scale. It is essential that this decay and in-growth chain be evaluated when looking at the RST. Since the half–lives of ^{237}Np and ^{233}U are long; the in-growth of ^{233}U and subsequent daughters can be effectively excluded in our 1,000-year decay and transport calculations.
  - $^{242}Pu \rightarrow {}^{238}U$: Because of the long half-life of ^{242}Pu the in-growth of ^{238}U from ^{242}Pu is not of concern in our 1,000-year time frame. Because ^{238}U and ^{234}U have relatively long half–lives, in-growth of ^{234}U and its further decay into additional daughters can be effectively excluded in our 1,000-year decay and transport calculations.

- $^{243}Am \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$: The combination of a relatively long half-life for ^{243}Am and a relatively small original source term compared to that of ^{239}Pu makes the in-growth of ^{239}Pu unimportant. Although ^{239}Pu has a long half-life relative to the 1000-year time period evaluated it has a large source term compared to ^{235}U. In-growth should be treated explicitly as it becomes significant just after the 1000-year time frame. Uranium-235 has a long half–life and further decay into additional daughters can be safely ignored in 1,000-year decay and transport calculations.
- $^{244}Cm \rightarrow {}^{240}Pu \rightarrow {}^{236}U \rightarrow {}^{232}Th$: The in-growth of ^{240}Pu should be treated explicitly although the contribution may be insignificant where the ^{240}Pu source term is much larger than ^{244}Cm. The in-growth of ^{236}U from ^{240}Pu must be treated explicitly. The long half-life of ^{236}U suggests that the in-growth of ^{232}Th from ^{236}U, and subsequent daughters will not be significant over the 1,000–year decay period.

It must be emphasized that conclusions drawn from this document are based on the 1000-year time frame and the same conclusions need not hold for either shorter or longer time periods.

1. Introduction

The radionuclides deposited by an underground nuclear test are derived from the original fuel material in the device not consumed in the explosion, nuclear reactions driving the explosion, and activation products created in the geologic medium. Collectively, these radionuclides and their residual abundances comprise the radionuclide source term or RST of the test. Following detonation, and depending on the presence of water at the location of the detonation, the residual radionuclides may be found in aqueous or gaseous states, precipitated or chemically sorbed states, or incorporated in melt glass produced by the test. The purpose of this report is to assess the decay and in-growth of radionuclides from the RST deposited by underground nuclear weapons tests conducted at the NTS from 1951 through 1992. The abundance of a radionuclide changes with time due to radioactive decay. The in-growth of radioactivity is the additional amount of radioactivity for a given radionuclide that comes from the decay of the parent isotopes. In this report, decay and in-growth of radionuclides from the RST are evaluated over the 1000-year time frame in order to determine whether coupled in-growth and decay affect the relative abundance of any RST radionuclide. It is also necessary to identify whether any new additional, derivative radionuclides resulting from in-growth need to be included in the radionuclide inventory.

One of the major goals of this report is to simplify the transport modeler's task by pointing out where in-growth is unimportant and where it needs to be considered. The specific goals of this document are to evaluate radionuclide decay chains and provide specific recommendations for incorporating radionuclide daughters of concern in the calculation of the radionuclide inventory. An unclassified inventory of radionuclides produced by 828 underground nuclear tests conducted at the NTS, subdivided into five principal geographic test areas, is presented in Bowen et al., (2001). The Yucca Flat geographic test area was further subdivided into tests below or within 100 m of the water table and those above. This radionuclide inventory by Bowen et al. (2001) is the data set from which we evaluate relevant decay and in-growth of radionuclides. This inventory provides a calculated estimate of radioactivity remaining underground at the NTS after underground nuclear testing ceased on Sept. 23, 1992.

The reasons for incorporation of decay chains in the assessment of the RST are:

(1) Several nuclides in the RST compiled by Lawrence Livermore (LLNL) and Los Alamos National Laboratories (LANL) (Bowen et al., 2001), radioactively decay to other radionuclides included in the inventory. Whereas the evolution of a radioactive parent is described by simple exponential decay; $N(t) = N(0)e^{-\lambda t}$ the evolution of radioactive daughters and granddaughters must include in-growth, resulting in coupled equations of the form $dN_2(t)/dt = N_1(t)\lambda_1 - N_2(t)\lambda_2$ where; $N_1 \xrightarrow{\lambda_1} N_2 \xrightarrow{\lambda_2}$, where N= number of atoms, t=time, λ=ln(2)/half-life.

(2) The decay of radionuclides may have geochemical implications for the migration of radionuclides away from underground nuclear test cavities. Modeling of the near-field for proposed high level waste repositories has shown that precipitation fronts may be induced by radionuclide decay and elemental solubility limits

(Worgan and Apted, 1992). Implications for far-field transport are also important. For example, in the case of a long-lived mobile parent decaying to a shorter-lived and less mobile daughter, the geochemical properties of the parent element may control the migration potential of the daughter nuclide.

(3) Finally, certain radionuclides regulated under Federal drinking water standards (Environmental Protection Agency, 1991) and not included in the RST (e.g. ^{222}Rn, ^{226}Ra, ^{228}Ra and a number of others) are produced by the radioactive decay of RST parent nuclides. Furthermore, gross alpha and beta activities in groundwater, which are also regulated under Federal drinking water standards (Environmental Protection Agency, 1991) represent the combined activities of all parent nuclides and their daughters.

2. Sources of Radionuclide Data

A priority of the Underground Test Area (UGTA) project, administered by the Environmental Restoration Division of NNSA/NV, was to determine as accurately as possible the total radionuclide inventory deposited in the subsurface at the NTS. The radionuclide inventory is a calculated estimate of the RST deposited underground resulting from nuclear testing and is a necessary starting point for estimations of radionuclide transport away from test locations. The initial effort to compile a total radionuclide inventory at the NTS used both measurements and calculations. The large amount of data required for this effort was collected from LLNL and LANL weapons program databases and was described in a preliminary classified report (Goishi et al., 1994). The final report was published in Miller et al. (2001). Because radionuclide data for individual nuclear tests are classified, the radionuclide inventory was subsequently divided into five geographic areas, summed for each radionuclide of concern in each geographic area, and published in an unclassified report (Bowen et al., 2001).

The five geographic areas used to sum the radionuclide abundances are shown in Table 1. The five areas are similar to the areas designated as "Corrective Action Units" for groundwater remediation.

TABLE 1
Principal Geographic Test Centers for NTS
(*from* Bowen et al., 2001)

NTS Principal Geographic Test Centers	NTS Areas Included
Yucca Flat*	1,2,3,4,6,7,8,9,10,15
Pahute Mesa – 19	19
Pahute Mesa – 20	20
Frenchman Flat	5,11
Rainier Mesa/Shoshone Mt	12,16,18,30

*Further subdivided into tests detonated above and below water table.

Because the data in the Bowen et al. (2001) report are the basis for our decay chain analysis in this report, it is important that we restate the criteria that were used to

determine which radionuclides to include in the radionuclide inventory. Not all radionuclides produced during a nuclear test are included in the radionuclide inventory. Some radionuclides have already decayed away; nuclides that have short half-lives decay to undetectable levels soon after the test. Other nuclides are produced in abundances too low to ever exceed levels deemed unsafe. For this reason the following criteria were used (Bowen et al., 2001):

(1) "Excluded were nuclides produced in such low amounts that if all of the amount produced during a nuclear test were dissolved into a volume of water equal to the volume of the detonation cavity for the test, 100 years from 1992 the resulting aqueous concentration (activity) in μCi/mL (one μCi = 2.22×10^6 disintegrations per minute) would be less than one-tenth of the values for the maximum permissible concentrations (MPC's) proposed for drinking water by the U. S. Environmental Protection Agency in the Federal Register (1991).

This effectively excludes almost all radionuclides with half- lives less than ten years.

(2) If a radionuclide exceeded criteria (1) for at least one test, it was included for all other tests for which estimates are available even if concentrations were below the 0.1 MPC criteria.

The MPC of a nuclide listed in this compilation is that concentration in drinking water that will impart a dose of 4 mrem/year to a person drinking an average of 2 liters of water per day. The requirement of 100 years into the future eliminates many nuclides that are produced in great abundance in nuclear detonations but have half-lives sufficiently short that they will have decayed below the 0.1 MPC value by that time.

(3) For nuclides with no listed values, we conservatively assumed a value of 10^{-8} Ci/mL for the MPC."

The 43 radionuclides that met the above criteria make up the radionuclide inventory listed in Table 1 in Bowen et al., (2001) and repeated here for completeness (Table 2). Although the radionuclide inventory includes activation products created in the host geologic medium, it does not include pre-existing, naturally occurring radionuclides, although they may become redistributed or incorporated in melt glass as a result of the test. In addition, it does not include any non-radioactive materials introduced underground by the test, even though many of these might also be considered contaminants.

TABLE 2
Radionuclides for Inclusion into Radionuclide Inventory
(*from* Bowen et al., 2001)

Element	Nuclide	Half-life (y)*	MPC (μCi/mL)	Main Source(s) (FP=fission product)
Hydrogen	^{3}H	12.32	6.1×10^{-5}	device component; ^{6}Li (n,α) T
Carbon	^{14}C	5715	3.2×10^{-6}	^{14}N (n,p); ^{13}C (n,γ); ^{17}O (n,α)

Aluminum	^{26}A1	7.1 x 10^{5}	----	^{27}A1 (n,2n)
Chlorine	^{36}C1	3.01 x 10^{5}	1.8 x 10^{-6}	^{35}C1 (n,γ); ^{39}K (n,α)
Argon	^{39}Ar	269	----	^{39}K (n,p); ^{38}Ar(n,γ)
Potassium	^{40}K	1.27 x 10^{9}	----	natural
Calcium	^{41}Ca	1.03 x 10^{5}	----	^{40}Ca (n,γ)
Nickel	^{59}Ni	7.6 x 10^{4}	2.7 x 10^{-5}	^{58}Ni (n,γ)
	^{63}Ni	100	9.9 x 10^{-6}	^{62}Ni (n,γ), ^{64}Ni (n,2n), ^{63}Cu (n,p)
Krypton	^{85}Kr	10.76	----	FP; ^{84}Kr (n,γ)
Strontium	^{90}Sr	28.78	4.2 x 10^{-8}	FP
Zirconium	^{93}Zr	1.5 x 10^{6}	5.1 x 10^{-6}	FP; ^{92}Zr (n,γ); ^{94}Zr (n,2n)
Niobium	^{93m}Nb	16.1	1.0 x 10^{-5}	^{93}Nb (n,n′)
	^{94}Nb	2.0 x 10^{4}	7.1 x 10^{-7}	FP; ^{93}Nb(n,γ)
Technetium	^{99}Tc	2.13 x 10^{5}	3.8 x 10^{-6}	FP; ^{99}Ru (n,p)
Palladium	^{107}Pd	6.5 x 10^{6}	3.7 x 10^{-5}	FP; ^{106}Pd (n,γ)
Cadmium	^{113m}Cd	14.1	----	FP
Tin	^{121m}Sn	~55	2.3 x 10^{-6}	FP; ^{120}Sn (n,γ)
	^{126}Sn	2.5 x 10^{5}	2.9 x 10^{-7}	FP
Iodine	^{129}I	1.57 x 10^{7}	2.1 x 10^{-8}	FP; ^{129}Xe (n,p)
Cesium	^{135}Cs	2.3 x 10^{6}	7.9 x 10^{-7}	FP
	^{137}Cs	30.07	1.2 x 10^{-7}	FP; ^{137}Ba (n,p)
Samarium	^{151}Sm	90	1.4 x 10^{-5}	FP; ^{150}Sm (n,γ)
Europium	^{150}Eu	36	----	^{151}Eu (n,2n)
	^{152}Eu	13.54	8.4 x 10^{-7}	^{151}Eu (n,γ); ^{153}Eu (n,2n)
	^{154}Eu	8.593	6.7 x 10^{-7}	^{153}Eu (n,γ)
Holmium	^{166}Ho	1.2 x 10^{3}	----	FP; ^{165}Ho (n,γ)
Thorium	^{232}Th	1.40 x 10^{10}	9.2 x 10^{-8}	natural and device component
Uranium	^{232}U	69.8	1.0 x 10^{-8}	device component; ^{233}U (n,2n)
	^{233}U	1.592 x 10^{5}	2.6 x 10^{-8}	device component; radiochemical tracer
	^{234}U	2.46 x 10^{5}	2.6 x 10^{-8}	natural and device component
	^{235}U	7.04 x 10^{8}	2.6 x 10^{-8}	natural and device component
	^{236}U	2.342 x 10^{7}	2.7 x 10^{-8}	device component; ^{235}U (n,γ); ^{238}U (n,2n)[2]
	^{238}U	4.47 x 10^{9}	2.6 x 10^{-8}	natural and device component
Neptunium	^{237}Np	2.14 x 10^{6}	7.2 x 10^{-9}	radiochemical tracer; decay of ^{237}U
Plutonium	^{238}Pu	87.7	7.2 x 10^{-9}	device component; radiochemical tracer; ^{239}Pu (n,2n); ^{237}Np (n,γ)
	^{239}Pu	2.410 x 10^{4}	6.5 x 10^{-8}	device component; decay of ^{239}U
	^{240}Pu	6.56 x 10^{3}	6.5 x 10^{-8}	device component; ^{239}Pu (n,γ); decay of ^{240}U
	^{241}Pu	14.4	----	device component; ^{240}Pu (n,γ); decay of ^{241}U
	^{242}Pu	3.75 x 10^{5}	6.8 x 10^{-8}	device component; radiochemical tracer; ^{241}Pu (n,γ); decay of ^{242}U
Americium	^{241}Am	432.7	6.4 x 10^{-9}	device component; radiochemical tracer; decay of ^{241}Pu
	^{243}Am	7.37 x 10^{3}	6.5 x 10^{-9}	device component; radiochemical

Curium	^{244}Cm	18.1	1.0×10^{-8}	tracer radiochemical tracer

* Half-lives obtained from GE Chart of the Nuclides, Fifteenth Edition (1996).

Prior to the release of the Bowen et al. (2001) report, several UGTA funded studies developed criteria for selecting radionuclides for application to specific transport modeling studies. Several reports used a different number of radionuclides of concern as well as slightly different numbers for the initial radioactivity. There are several reasons for this *apparent* lack of agreement. Some works, such as Goishi et al., (1994), Esser (1994), Smith (2001) and Pawloski et al., (2002) used curie activities and atoms decay corrected to January 1, 1994. Other works such as Miller et al. (2001) and Bowen et al. (2001), decay corrected the inventory to September 23, 1992, the last date of underground nuclear testing at the NTS. In addition, Smith (2001) used the Department of Energy classification bulletin WNP-87 prior to the release of Bowen et al. (2001) to calculate the radionuclide inventory for all tests fired below or within 100 m of the water table in Areas 19 and 20 (Pahute Mesa). Pawloski et al. (2002) used the Smith (2001) compilation for the radionuclide inventory of the Cheshire test in Pahute Mesa. The Bowen et al. (2001) inventory did not separate the Pahute Mesa radionuclide inventory for tests detonated above and below the water table as the Smith (2001) report did. Tompson et al., (2002) used the Bowen et al. (2001) inventory for calculation of the hydrologic source term in Frenchman Flat.

In addition to reports using slightly different dates to decay correct the atoms in the RST, different numbers of radionuclides of concern were also used. Again, this *apparent* lack of agreement is mainly a result of different criteria and goals for the various projects. Smith (2001) used the same 43 radionuclides as Bowen et al. (2001), but split Th and U into soil and device contributions. He also footnoted the possible addition of ^{150}Gd, and provided a summed inventory for ^{150}Gd that was deposited below or within 100 m of the water table. Tompson et al. (2002) used the 43 radionuclides from Bowen et al. (2001) but added ^{150}Gd and ^{152}Gd. There is no initial abundance of ^{150}Gd as it is solely derived from the decay of ^{150}Eu. ^{152}Gd is produced from the decay of ^{152}Eu and also has no initial abundance. Pawloski et al. (2002) used the radionuclide inventory of Smith (2001) plus additional criteria to initially evaluate 52 radionuclides of concern for the Cheshire test. Although initially discussed, eight species for which groundwater measurements have been reported were ultimately not included Cheshire inventory because there was no reported radionuclide inventory because they do not satisfy criterion (1) in Bowen et al. (2001). All have half-lives less than ten years; they are ^{22}Na, ^{54}Mn, ^{60}Co, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{144}Ce, and ^{155}Eu.

We included this account of the different strategies studied to select radionuclides of concern at the NTS to emphasize that initial assumptions must be evaluated and care should be exercised before comparing radionuclide inventories used in previous reports. For this report, we adopt the values given in Bowen et al. (2001) as our standard.

3. Criteria for Recommendations

The principal goal of this report is to simplify the transport modeler's task by pointing out where in-growth is unimportant and where it needs to be considered. For the RST, assessing the importance of the decay and in-growth is relatively straightforward. Uncertainties are limited to the accuracy of the radionuclide inventory. Bowen et al. (2001) estimated uncertainties for the various groups of radionuclides in the radionuclide inventory and the table is reproduced below (Table 3). Uncertainties in the regional actinide inventories are thought to be less than 20%, and will not have a significant effect on the evolution curves and the recommendations presented in this report.

TABLE 3
Estimated Accuracies for Individual Nuclides in
the Various Groups of Radionuclides
(*from* Bowen et al., 2001)

Fission products:	~ 10 to 30 % for most of the fission products
Unspent fuel materials:	~ 20 % or better
Fuel activation products:	~ 50 % or better
Residual tritium:	~ 300% or better
Activation products:	~ a factor of 10

4. Systematics of Radioactive Decay and RST Evolution

The systematics of radioactive decay in a closed system are well known (Friedlander et al., 1981). The generalized Bateman's equation for the decay chain

$$N_1 \xrightarrow{\lambda_1} N_2 \xrightarrow{\lambda_2} \ldots N_i \xrightarrow{\lambda_i} N_{i+1} \xrightarrow{\lambda_{i+1}} \ldots N_n \xrightarrow{\lambda_n}$$

where daughter initial activities are nonzero is

$$\begin{aligned} N_n(t) = N_1(0)\left\{C_{11}e^{-\lambda_1 t} + C_{12}e^{-\lambda_2 t} \ldots + C_{1j}e^{-\lambda_j t} \ldots + C_{1n}e^{-\lambda_n t}\right\} \\ +\ldots \\ +N_i(0)\left\{C_{ii}e^{-\lambda_i t} + C_{i(i+1)}e^{-\lambda_{i+1} t} \ldots + C_{ij}e^{-\lambda_j t} \ldots + C_{in}e^{-\lambda_n t}\right\} \\ +\ldots \\ +N_n(0)C_{nn}e^{-\lambda_n t} \end{aligned} \qquad (1)$$

where $C_{ij} = \dfrac{\lambda_i \lambda_{i+1} \ldots \lambda_{n-1}}{\left(\lambda_i - \lambda_j\right)\left(\lambda_{i+1} - \lambda_j\right)\ldots\left(\lambda_n - \lambda_j\right)}$

$N = atoms, t = time$

and $\lambda = \ln(2)/half - life$

Branching decay is handled by introducing a branching fraction. For the case where $N_i \xrightarrow{f_i} N_{i+1}$ and f is a fraction between 0 and 1, $f_i\lambda_i$ replaces λ_i in the numerator of all decay-constant coefficients where λ_i appears, e.g.

$$C_{ij} = \frac{f_i\lambda_i\lambda_{i+1}\ldots\lambda_{n-1}}{\left(\lambda_i - \lambda_j\right)\left(\lambda_{i+1} - \lambda_j\right)\ldots\left(\lambda_n - \lambda_j\right)}$$

In our application of Bateman's equations to radionuclide inventory nuclides and their daughters, we assumed that all daughter nuclides with half-lives of less than one year were in secular equilibrium with their parent nuclides. Secular equilibrium is a special condition that occurs for $N_1 \xrightarrow{\lambda_1} N_2 \xrightarrow{\lambda_2}$ when $\lambda_2 >> \lambda_1$. After several daughter half-lives (λ_2), the activity of the daughter will be equal to the activity of the parent, i.e. $\lambda_2 N_2 = \lambda_1 N_1$. Once secular equilibrium is reached, the daughter may be eliminated from the decay chain in Bateman's equations, i.e. $N_1 \xrightarrow{\lambda_1} N_2 \xrightarrow{\lambda_2} N_3$ becomes $N_1 \xrightarrow{\lambda_1} N_3$.

The final complication is nuclides with more than one parent. For example, the decay of ^{238}U (though short-lived intermediaries) *and* ^{238}Pu decay to ^{234}U, or the decay of ^{232}U *and* ^{228}Ra to ^{228}Th. These cases result in an additional term in the Bateman's equation, e.g.

$$\ldots + N_{1a}(0)\left\{C_{1a1a}e^{-\lambda_{1a}t} + C_{1a2}e^{-\lambda_2 t}\ldots + C_{1aj}e^{-\lambda_j t}\ldots + C_{1an}e^{-\lambda_n t}\right\}\ldots$$

All the species in a single decay chain constitute a radioactive family or series. Many of the radionuclides in the RST have simple, fairly short decay chains. All of the radionuclides listed in the Bowen et al. (2001) inventory that have a high atomic number (Z>80) have long, complex decay chains (e.g. Z_{Th}=90). There are four important series that include the majority of nuclides listed in the radionuclide inventory with Z>80 (Figures 1-4). Figures 1-4 depict the decay relationships between the radionuclide chains for the four actinide series (from Friedlander et al. (1981)). Although not depicted in Figures 1-4, the different isotopes of Pu, Am and Cm decay into these four chains and can be easily followed by using equations 9-13. For example, ^{241}Pu decays to ^{241}Am, which decays to ^{237}Np in the neptunium series (equation 10, figure 2). Uranium-238 is the parent in the uranium series, a 14-transformation decay chain that ultimately decays to the stable nuclide, ^{206}Pb (Figure 1). Neptunium-237 is the parent to the neptunium series that decays to the stable nuclide, ^{209}Bi (Figure 2). The 4n + 3, or actinium series has ^{235}U as the parent and ^{207}Pb is the stable end member (Figure 3). Thorium (^{232}Th) is the parent of the 4n or thorium series and it decays to the stable nuclide, ^{208}Pb (Figure 4).

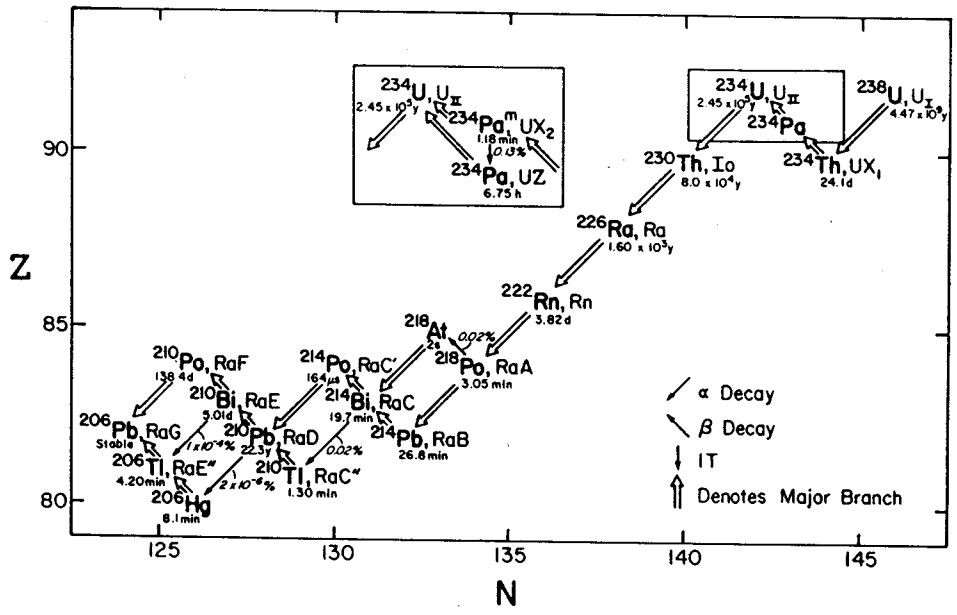

Figure 1. The uranium (U) decay series.

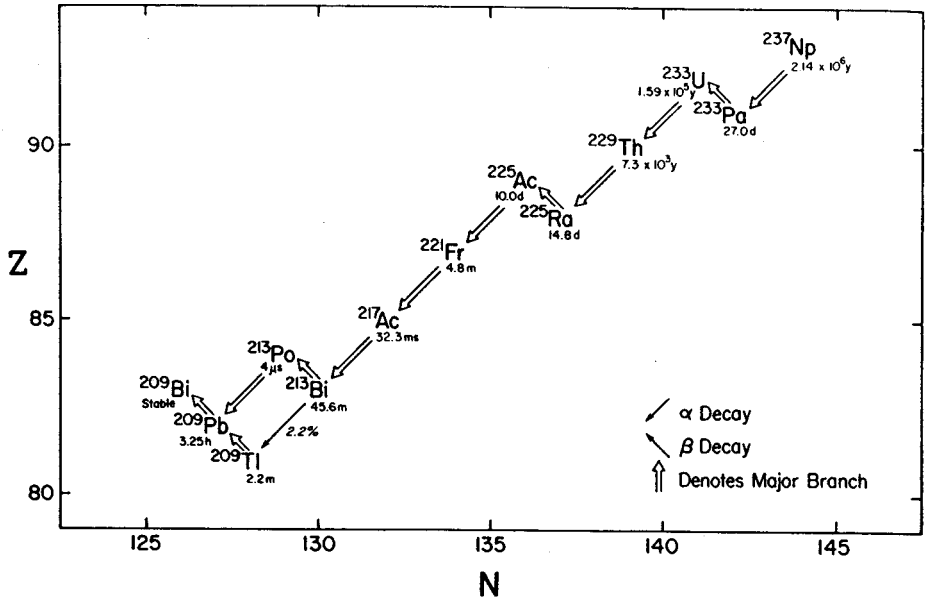

Figure 2. The neptunium (Np) decay series.

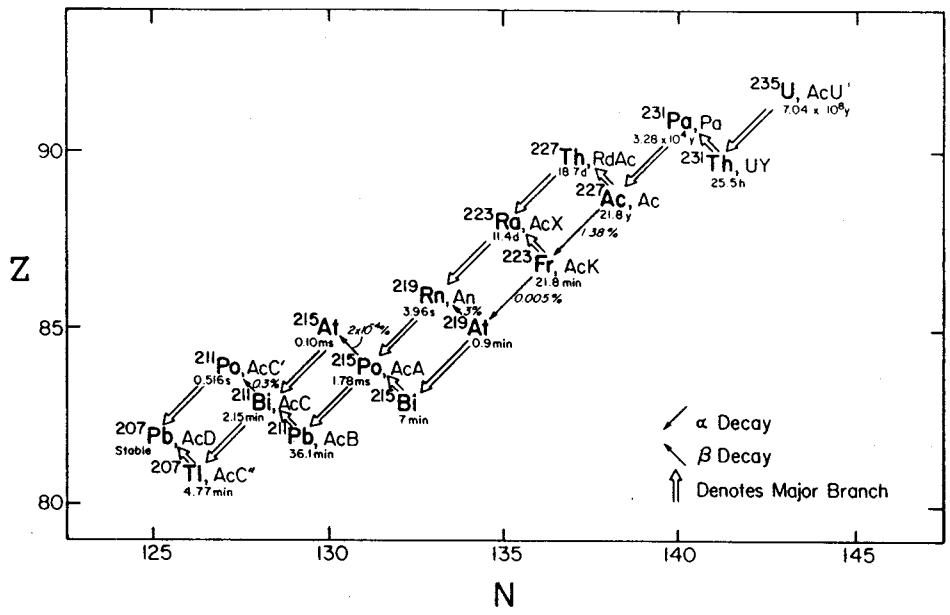

Figure 3. The actinium (Ac) decay series.

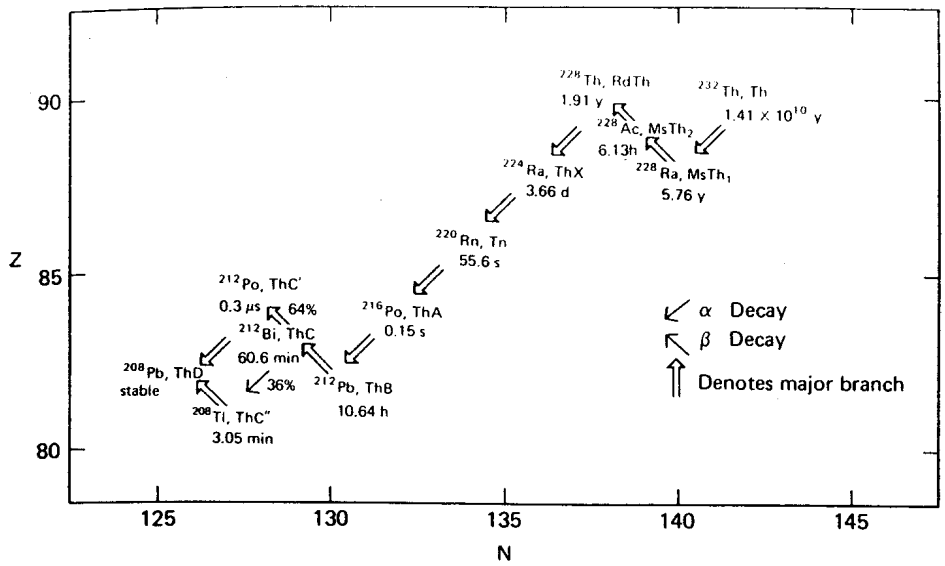

Figure 4. The thorium (Th) decay series.

5. Decay Chains Considered

Microsoft Excel© spreadsheets were used to model the evolution of each radionuclide in the inventory and their daughters from 0–1,000,000 years. Bateman's equations were used with modifications as necessary to account for branching and nuclides with more than one parent. Only nuclides with half-lives of greater than one year were explicitly considered; intermediaries with half-lives of less than one year were considered to be in secular equilibrium with their parents allowing for the simplification of the equations. Time zero was September 23, 1992, and initial inventories were for the five geographic areas designated in Bowen et al. (2001). The time steps were logarithmic, i.e. $0,1,2,\ldots,10,20,\ldots,100,200,\ldots$. A closed system was assumed, i.e. the evolution of the total radioactive inventory integrated over all phases and spatial scales was modeled.

A selection of results from modeling the evolution of each radionuclide in the inventory is shown in Figures 5 through 8. Figures 5 through 8 show the activity (Ci) for each radionuclide plotted against time for all the radionuclides that decay into the four most important and complex decay series (Figures 1-4). The figures are plotted for each of the five geographic divisions in the radionuclide inventory. The figures clearly show the effects of radioactive decay with time for individual radionuclides and the relative contribution of in-growth of radioactivity from parent isotopes.

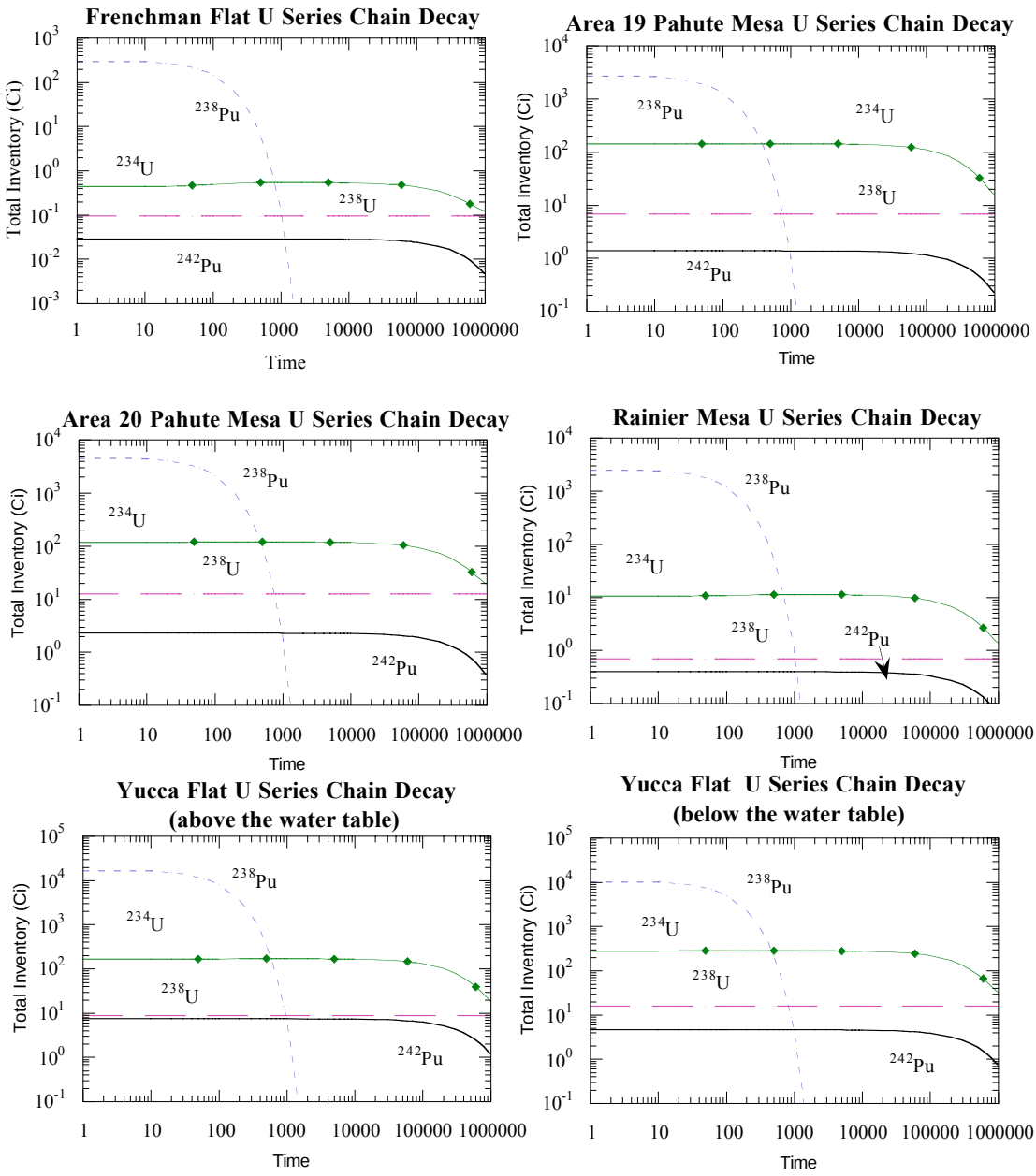

Figure 5. The activity of the each radionuclide of concern in the U-series is plotted vs. time for each of the five geographic areas designated by Bowen et al. (2001).

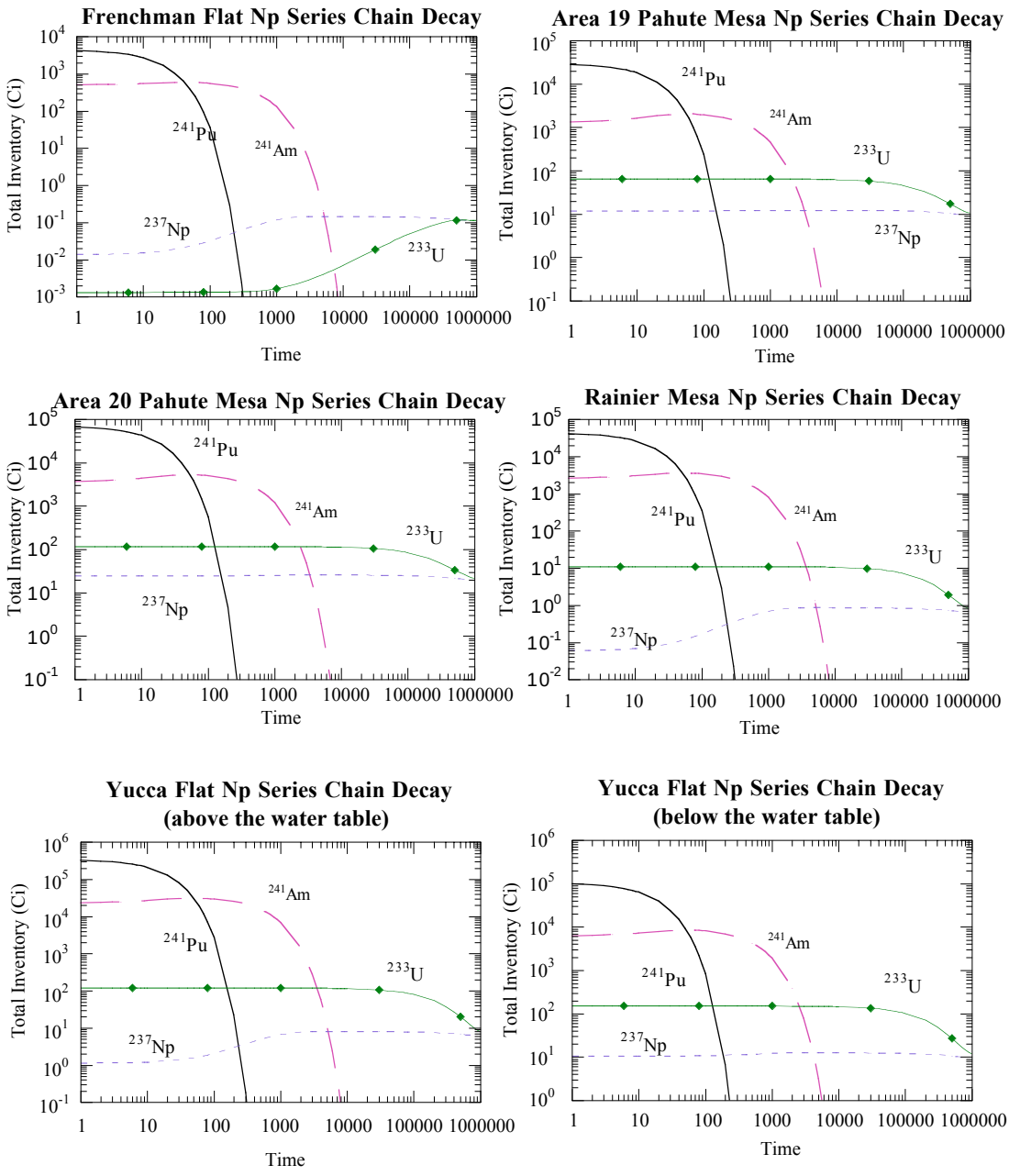

Figure 6. The activity of the each radionuclide of concern in the Np-series is plotted vs. time for each of the five geographic areas designated by Bowen et al. (2001).

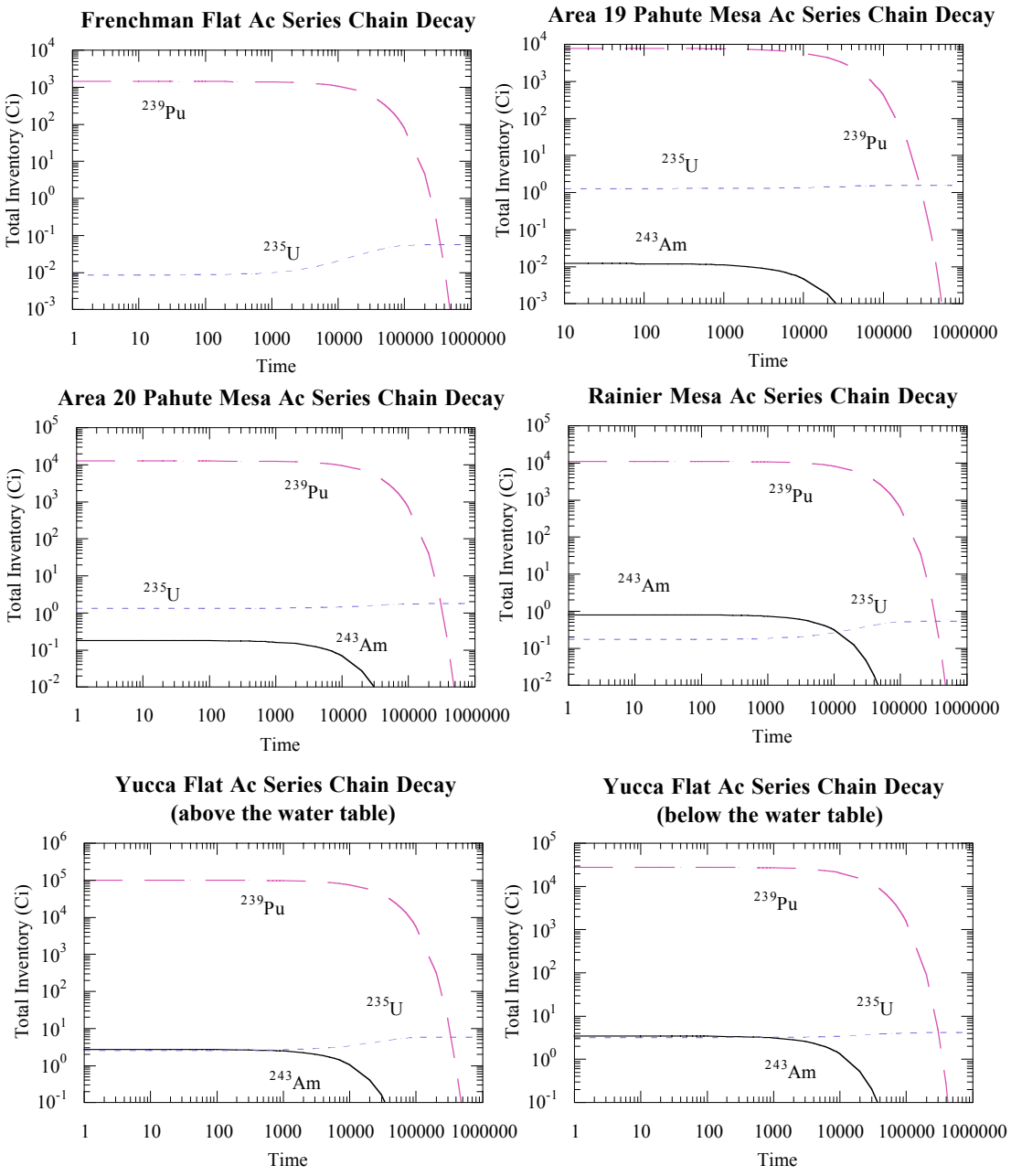

Figure 7. The activity of the each radionuclide of concern in the Ac-series is plotted vs. time for each of the five geographic areas designated by Bowen et al. (2001).

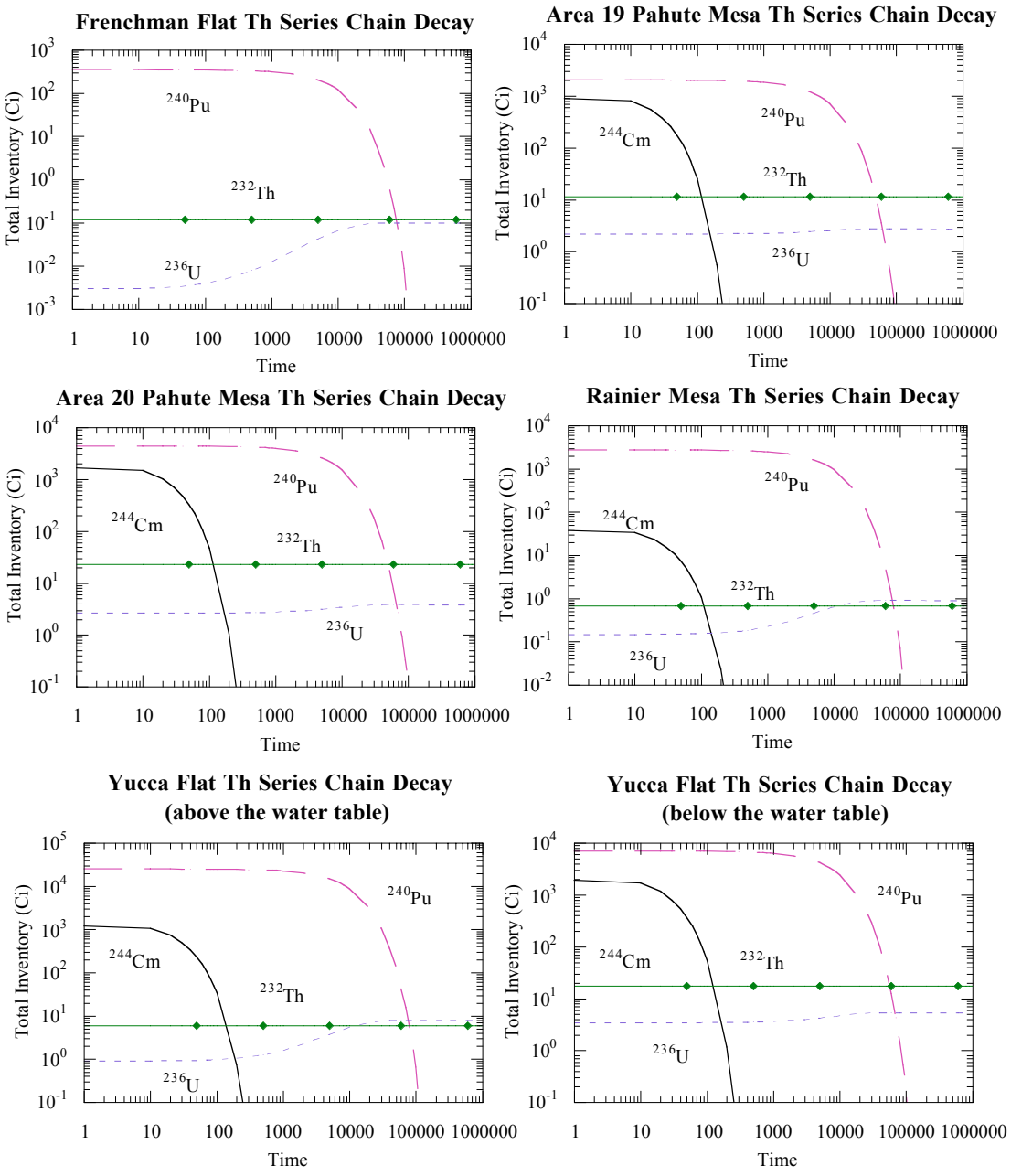

Figure 8. The activity of the each radionuclide of concern in the Th-series is plotted vs. time for each of the five geographic areas designated by Bowen et al. (2001).

5.1 General Observations from the Radionuclide Inventory

By definition, all of the 43 radionuclides selected for inclusion in the radionuclide inventory will undergo radioactive decay. After careful review of all 43 radionuclides the following observations were made:

(1) All of the radionuclides in the radionuclide inventory with low to intermediate atomic numbers (Z<80) will decay directly into stable, nonradioactive daughter products. *They will not produce additional, derivative members of the radionuclide inventory*. These 19 radionuclides from the radionuclide inventory are: ^{3}H, ^{14}C, ^{26}Al, ^{36}Cl, ^{39}Ar, ^{41}Ca, ^{59}Ni, ^{63}Ni, ^{85}Kr, ^{94}Nb, ^{99}Tc, ^{107}Pd, ^{113m}Cd, ^{121m}Sn, ^{129}I, ^{135}Cs, ^{151}Sm, ^{154}Eu, ^{166}Ho.

(2) Because ^{40}K is overwhelmingly of natural origin, it will not be considered further as a component of the RST.

(3) Four members of the radionuclide inventory (^{90}Sr, ^{126}Sn, ^{137}Cs, ^{232}U) decay to short-lived radioactive daughter products with $t_{1/2}$< 1 week, which in turn, decay into stable granddaughters (see below, equations 2-5). In general, all short-lived intermediaries will exist in secular equilibrium. Their abundance will be smaller than, and can be calculated as an instantaneous function of, their parent's abundance. *Accordingly, inclusion of the daughters as additional, derivative members of the radionuclide inventory is not necessary.* Further discussion follows in Section 5.2.

(4) All of the radionuclides with high atomic number (Z>80) in the radionuclide inventory are members of decay chains that include two or more members of the radionuclide inventory. These 19 radionuclides are members of eight additional decay chains. They are: ^{93}Zr, ^{93}Nb, ^{150}Eu, ^{152}Eu, ^{232}Th, ^{233}U, ^{234}U, ^{234}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, and ^{244}Cm.

The eight decay chains will be discussed further in Section 5.3 in terms of the relevant decay and in-growth that need to be addressed over a 1,000-year period.

(5) There are daughters in the actinide chains (Figures 1-4), such as isotopes of radon and radium, which are regulated under Federal drinking water standards but not included in the radionuclide inventory. In all cases, the inventories are dominated by the natural component. For example, as the initial ^{226}Ra (device + natural) moves out of the cavity, it will interact with a larger volume of rock containing significant U and Ra, and the device component will decrease according to the inverse square law. *Therefore, they do not need to be included as additional derivative members of the radionuclide inventory.*

5.2 Four Radionuclides that Decay to Short-Lived Nuclides: ^{90}Sr, ^{26}Sn, ^{137}Cs, and ^{232}U

The following four chains (paragraph 3 above) are characterized by a parent radionuclide in the radionuclide inventory decaying into a stable, end-member daughter via one or more short-lived intermediary radionuclides.

$$^{90}\mathrm{Sr} \xrightarrow{29.1y/\beta} {}^{90}\mathrm{Y} \xrightarrow{64h/\beta} {}^{93}\mathrm{Zr(stable)} \quad (2)$$

$$^{126}\mathrm{Sn} \xrightarrow{\sim 10^{6}y/\beta} {}^{126}\mathrm{Sb} \xrightarrow{12.5d/\beta} {}^{126}\mathrm{Te(stable)} \quad (3)$$

$$^{137}\mathrm{Cs} \xrightarrow{30.1y/\gamma} {}^{137\mathrm{m}}\mathrm{Ba} \xrightarrow{2.5m/IT} {}^{137}\mathrm{Ba(stable)} \quad (4)$$

$$^{232}\mathrm{U} \xrightarrow{70y/\alpha} {}^{228}\mathrm{Th} \xrightarrow{1.9y/\alpha} 7 \text{ short-lived intermediaries...} \longrightarrow {}^{208}\mathrm{Pb(stable)} \quad (5)$$

Strontium-90, ^{126}Sn and ^{137}Cs (equations 2-4) all have short decay chains; however ^{232}U decays to a long, 10-member chain finally decaying to ^{208}Pb, its stable end-member daughter (equation 5, Figure 4). ^{232}U decays to short-lived ^{228}Th that has a half-life of 1.9 years and decays via alpha-emission into several short-lived intermediaries whose half–lives range from microseconds to 3.6 days. Of these, four (^{224}Ra, ^{220}Rn, ^{216}Po, and ^{212}Po) specifically decay via alpha-emission. However, because the half-lives of the ^{232}U daughters are short, we can assume they all exist in secular equilibrium with ^{232}U. Therefore, their abundances can be estimated as an instantaneous function of their parent's abundance. Inclusion of the daughters as additional, derivative members of the radionuclide inventory is not necessary.

5.3 Eight Decay Chains Involving Radionuclide Inventory Nuclides

^{93}Zr chain. Both ^{93}Zr and ^{93m}Nb are members of the radionuclide inventory. Because of the long half–life of ^{93}Zr the in-growth of ^{93m}Nb from this decay chain (equation 6) over 1,000 years should not be significant, so both can be treated independently in decay and transport calculations.

$$^{93}\mathrm{Zr} \xrightarrow{\sim 10^{6}y/\beta} {}^{93m}\mathrm{Nb} \xrightarrow{16.1y/IT} {}^{93}\mathrm{Nb\ (stable)} \quad (6)$$

^{150}Eu chain. Eu-150 has a relatively short half-life so that over the 1000-year time frame all of the ^{150}Eu will decay to ^{150}Gd. Gd-150, is an additional, derivative radionuclide that was not included in the radionuclide inventory of Bowen et al. (2001) because it is not initially produced by a nuclear test. Its inventory can be estimated from the decay of ^{150}Eu. Gd-150 has such a long half-life, that over a 1,000-year analysis the daughter radionuclides can be effectively ignored in 1000-year decay and transport calculations.

$$^{150}\mathrm{Eu} \xrightarrow{36y/\beta} {}^{150}\mathrm{Gd} \xrightarrow{\sim 10^{6}y/\alpha} {}^{146}\mathrm{Sm} \xrightarrow{\sim 10^{8}y/\alpha} {}^{142}\mathrm{Nd\ (stable)} \quad (7)$$

^{152}Eu chain. Eu-152 has a relatively short half-life so that over the 1000-year time frame all of the ^{152}Eu will decay to ^{152}Gd. Gd-152, is an additional, derivative radionuclide that was not included in the radionuclide inventory of Bowen et al. (2001) because it is not initially produced by a nuclear test. Its inventory can be estimated from the in-growth from ^{152}Eu. Eu-152 decays in a 72% branch to ^{152}Sm (stable) and in a 28% branch to ^{152}Gd. Gd-152 has such a long half-life, that over a 1,000-year analysis daughter radionuclides of ^{152}Gd can be effectively ignored in 1000-year decay and transport calculations.

$$^{152}\mathrm{Eu} \xrightarrow{(28\%\ \text{branch})\ 13.5y/\beta} {}^{152}\mathrm{Gd} \xrightarrow{\sim 10^{14}\ y/\alpha} \ldots \qquad (8)$$

Subsequent daughter products produced beyond ^{152}Gd are ^{148}Sm (~10^{14} y half-life/α), ^{144}Nd (~10^{15} y/α), and ^{140}Ce (stable).

^{238}Pu chain. Both ^{238}Pu and ^{234}U are members of the radionuclide inventory and their abundance as a function of time has been modeled and plotted in Figure 5. The ^{238}Pu decays with a relatively short half–life into ^{234}U. The in-growth of ^{234}U will add to the inventory of ^{234}U from decay of ^{238}Pu. U-234 has a long half-life such that over the 1000-year time frame daughter products can be effectively ignored in 1,000-year decay and transport calculations.

$$^{238}\mathrm{Pu} \xrightarrow{87.7y/\alpha} {}^{234}\mathrm{U} \xrightarrow{\sim 10^{5}\ y/\alpha} \ldots {}^{206}\mathrm{Pb(stable)} \qquad (9)$$

^{241}Pu chain. Four of the first five members of decay chain (equation 10) are included as members of the radionuclide inventory and their abundance as a function of time has been modeled and plotted in Figure 6. The coupled decay and in-growth between ^{241}Pu, ^{241}Am, and ^{237}Np must be treated explicitly in a transport model because of their relatively short half-lives. The ^{241}Pu ($t_{1/2}$ = 14.4 y) will significantly increase the activity of the ^{241}Am and consequently the ^{237}Np on the 1000-year time scale. The in-growth of ^{241}Am can be observed by the slight increase in ^{241}Am curve at less than 100 years (Figure 6). Since the half–lives of ^{237}Np and ^{233}U are long, the in-growth of ^{233}U and subsequent daughters can be effectively excluded in our 1,000-year decay and transport calculations.

$$^{241}\mathrm{Pu} \xrightarrow{14.4y/b} {}^{241}\mathrm{Am} \xrightarrow{433y/a} {}^{237}\mathrm{Np} \xrightarrow{\sim 10^{6}\ y/a} {}^{233}\mathrm{Pa} \xrightarrow{\sim 27d/b} {}^{233}\mathrm{U} \xrightarrow{\sim 10^{5}\ y/a} \ldots {}^{209}\mathrm{Bi(stable)} \qquad (10)$$

^{242}Pu chain. Three members (^{242}Pu, ^{238}U, ^{234}U) in this decay chain (equation 11) are members of the radionuclide inventory and their abundance as a function of time has been modeled and plotted in Figure 5. Because of the long half-life of ^{242}Pu the in-growth of ^{238}U from ^{242}Pu is not of concern in our 1,000-year time frame. Because ^{238}U and ^{234}U

have relatively long half–lives, in-growth of ^{234}U and its further decay into additional daughters can be effectively excluded in our 1,000-year decay and transport calculations.

$$^{242}\mathrm{Pu} \xrightarrow{\sim 10^5\, y/\alpha} {}^{238}\mathrm{U} \xrightarrow{\sim 10^9\, y/\alpha} {}^{234}\mathrm{Th} \xrightarrow{\sim 24\, d/\beta} {}^{234}\mathrm{Pa} \xrightarrow{\sim 6\, h/\beta} {}^{234}\mathrm{U} \xrightarrow{\sim 10^5\, y/\alpha} \ldots {}^{206}\mathrm{Pb(stable)} \quad (11)$$

^{243}Am chain. Three members shown in decay chain (12) are members of the radionuclide inventory and their abundance as a function of time has been modeled and plotted in Figure 7. The combination of a relatively long half-life for ^{243}Am and a relatively small original source term compared to that of ^{239}Pu makes the in-growth of ^{239}Pu unimportant. Plutonium-239 has a long half-life relative to the 1000-year time period evaluated for this report so in-growth of ^{235}U is probably not important. In-growth would be important for time frames just greater than 1000 years, especially for areas where the original source term for ^{235}U is much lower than ^{239}Pu. Figure 7 shows the effects of in-growth of ^{235}U becoming important just after 1000 years. Uranium-235 has a long half–life and further decay into additional daughters can be safely ignored in 1,000-year decay and transport calculations.

$$^{243}\mathrm{Am} \xrightarrow{7370\, y/\alpha} {}^{239}\mathrm{Np} \xrightarrow{\sim 2\, d/\alpha} {}^{239}\mathrm{Pu} \xrightarrow{\sim 10^4\, y/\alpha} {}^{235}\mathrm{U} \xrightarrow{\sim 10^8\, y/\alpha} \ldots {}^{207}\mathrm{Pb(stable)} \quad (12)$$

^{244}Cm chain. Each of the first four members shown in decay chain (13) is a member of the radionuclide inventory and their abundance as a function of time has been modeled and plotted in Figure 8. Cm-244 has a short half-life and it will all decay to ^{240}Pu in the 1000-year time frame. The in-growth of ^{240}Pu should be treated explicitly although the contribution may be insignificant where the ^{240}Pu source term is much larger than ^{244}Cm. The in-growth of ^{236}U from^{240}Pu must be treated explicitly. The long half-life of ^{236}U suggests that the in-growth of ^{232}Th from ^{236}U, and subsequent daughters will not be significant over the 1,000–year decay period.

$$^{244}\mathrm{Cm} \xrightarrow{18.1 y/\alpha} {}^{240}\mathrm{Pu} \xrightarrow{6560 y/\alpha} {}^{236}\mathrm{U} \xrightarrow{\sim 10^7\, y/\alpha} {}^{232}\mathrm{Th} \xrightarrow{\sim 10^{10}\, y/\alpha} \ldots {}^{208}\mathrm{Pb(stable)} \quad (13)$$

6. Summary

The relevant decay and in-growth of radionuclides from the radiologic inventory compiled by Bowen et al. (2001) were evaluated over the 1000-year time frame in order to determine whether coupled in-growth and decay affect the relative abundance of any RST radionuclide. The radionuclide inventory data in Bowen et al. (2001) was the basis for our assessment as individual radionuclide inventory data for specific nuclear tests are classified.

All of the radionuclides in the radionuclide inventory with low to intermediate atomic numbers ($Z<80$) will decay directly into stable, nonradioactive daughter products. They will not produce additional, derivative members of the radionuclide inventory.

The short-lived radionuclides in decay chains with long-lived parents will exist in secular equilibrium with their parents and their abundances can be estimated from the abundance of their parent. Some examples of this are ^{90}Sr and ^{90}Y, ^{137}Cs and ^{137m}Ba, and ^{232}U and ^{228}Th. Inclusion of the daughters as additional, derivative members of the radionuclide inventory is not necessary.

There are daughters in the actinide chains, such as isotopes of radium and radon that are of concern in the environment, but the background from the natural uranium dwarfs the RST concentrations. Therefore, they do not need to be included as additional derivative members of the radionuclide inventory.

There are eight decay chains that involve long decay and in-growth patterns that have been discussed in this report.

1 ^{93}Zr → ^{93m}Nb → ^{93}Nb: The in-growth of ^{93m}Nb from ^{93}Zr over 1,000 years will not be significant. Over a 1000 year time frame all of the ^{93m}Nb will have decayed to stable ^{93}Nb, and therefore is not an additional, derivative radionuclide.

2 ^{150}Eu → ^{150}Gd → ^{146}Sm: Over the 1000 year-time frame all of the ^{150}Eu will decay to ^{150}Gd. ^{150}Gd is an additional derivative radionuclide to the radionuclide inventory of Bowen et al. (2001). Gd-150 has such a long half-life, that over a 1,000-year analysis the daughter radionuclides can be effectively ignored in 1000-year decay and transport calculations.

3 ^{152}Eu → ^{152}Gd: Over the 1000 year-time frame a fraction of the ^{152}Eu will decay to ^{152}Gd. ^{152}Gd is an additional derivative radionuclide to the radionuclide inventory of Bowen et al. (2001). Gd-152 has such a long half-life, that over a 1,000-year analysis the daughter radionuclides can be effectively ignored in 1000-year decay and transport calculations.

4 ^{238}Pu → ^{234}U: The ^{238}Pu will gradually decay to ^{234}U in the 1000 years. This in-growth of ^{234}U will add to the ^{234}U from decay of ^{238}Pu and should be treated explicitly. This may be of consequence in tests with little ^{234}U. Further daughter products of ^{234}U should be insignificant over 1000 years and do not represent additional, derivative members of the radionuclide inventory.

5 ^{241}Pu → ^{241}Am → ^{237}Np: This is probably the most important decay and in-growth chain in the RST. In-growth is important for calculating both ^{241}Am and ^{237}Np. The ^{241}Pu ($t_{1/2}$ = 14.4 y) will significantly increase the activity of the ^{241}Am and consequently the ^{237}Np on the 1000-year time scale. It is essential that this decay and in-growth chain be evaluated when looking at the RST. Since the half–lives of ^{237}Np and ^{233}U are long, the in-growth of ^{233}U and subsequent daughters can be effectively excluded in our 1,000-year decay and transport calculations.

6 ^{242}Pu → ^{238}U: Because of the long half-life of ^{242}Pu the in-growth of ^{238}U from ^{242}Pu is not of concern in our 1,000-year time frame. Because ^{238}U and ^{234}U have relatively long half–lives, in-growth of ^{234}U and its further decay into

additional daughters can be effectively excluded in our 1,000-year decay and transport calculations.

7 $^{243}Am \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$: The combination of a relatively long half-life for ^{243}Am and a relatively small original source term compared to that of ^{239}Pu makes the in-growth of ^{239}Pu unimportant. Although ^{239}Pu has a long half-life relative to the 1000-year time period evaluated it has a large source term compared to ^{235}U. In-growth should be treated explicitly as it becomes significant just after the 1000-year time frame. Uranium-235 has a long half–life and further decay into additional daughters can be safely ignored in 1,000-year decay and transport calculations.

8 $^{244}Cm \rightarrow {}^{240}Pu \rightarrow {}^{236}U \rightarrow {}^{232}Th$: The in-growth of ^{240}Pu should be treated explicitly although the contribution may be insignificant where the ^{240}Pu source term is much larger than ^{244}Cm. The in-growth of ^{236}U from ^{240}Pu must be treated explicitly. The long half-life of ^{236}U suggests that the in-growth of ^{232}Th from ^{236}U, and subsequent daughters will not be significant over the 1,000–year decay period.

It must be emphasized that conclusions drawn from this document are based on the 1000-year time frame and the same conclusions need not hold for either shorter or longer time periods. As shown in Figures 5 through 8, decay chains will vary with the time period of interest.

Acknowledgments

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